

Separate H₂ and CO production from CH₄-CO₂ cycling of Fe-Ni

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November 20, 2021

Abstract

Fe-Ni materials with varying Ni loading are developed for separate H₂ and CO production by CH₄-CO₂ chemical looping. The product streams are obtained by first feeding CH₄, which decomposes to H₂ and carbon. The latter acts as reductant for the subsequent CO₂ feed, which together with Fe re-oxidation yields CO. After 25 CH₄-CO₂ cycles, 10Fe₅Ni@Zr has a higher H₂ space-time-yield than 10Fe₀Ni@Zr ($20 \text{ mmol}[\text{s}]^{-1} \text{ kg}^{-1}(\text{Fe}+\text{Ni})^{-1}$ vs. $15 \text{ mmol}[\text{s}]^{-1} \text{ kg}^{-1}(\text{Fe}+\text{Ni})^{-1}$), a 2.6 times higher CO ($57 \text{ mmol}[\text{s}]^{-1} \text{ kg}^{-1}(\text{Fe}+\text{Ni})^{-1}$) and lower deactivation. This improvement has two reasons: (i) CH₄ activation over Ni leading to cracking, (ii) product hydrogen causing deeper FeO reduction. Deactivation follows from accumulated carbon, non-reactive for CO₂. On Ni and Fe sites, carbon can be removed by lattice oxygen or CO₂, yielding more CO compared to the theoretical value for Fe oxidation. However, carbon that migrates away from the metals requires oxygen for removal, which restores the activity of the Ni-containing samples.

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